IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Attorney Docket No.: UDL1P065

First Named Inventor: PARVIN

TRANSMITTAL LETTER FOR A PCT INTERNATIONAL APPLICATION ENTERING THE NATIONAL STAGE IN THE U.S. AS A DESIGNATED or ELECTED OFFICE UNDER 35 USC 371

Commissioner for Patents
Box PCT
Attention: DO/EO
Washington, DC 20231

    Transmitted herewith are the papers required to enter the national stage in the U.S. as a designated office/elected office for the following PCT international patent application:

INTERNATIONAL APPLICATION NUMBER: PCT/GB00/00009
Int'l Filing Date: **11 January 2000**
1st Priority Date: **11 January 1999**
Inventor(s):   **PARVIN, Daniel Francis; RONALDSON, John Paul**
For: **APPARATUS AND METHODS FOR INVESTIGATION OF RADIOACTIVE SOURCES IN A SAMPLE**

The United States Patent Office is: (select one)

☐ A Designated Office (No Demand was filed - See 37 CFR 1.494)

☒ An Elected Office (A Demand for Preliminary Examination was Filed - See 37 CFR 1.495)

Enclosures:

☒ A copy of the international application (if this line is not checked, the international application was previously communicated by the International Bureau or the international application was originally filed in the USPTO).

☐ An English Translation of the International Application

☐ A Combined Declaration and Power of Attorney

☐ A copy of amendments made under PCT Article 19

☐ A translation of amendments made under PCT Article 19

☐ A translation of amendments made under PCT Article 34 (annexes to the international preliminary examination report)

☐ Information Disclosure Statement

☐ An Assignment of the Invention to:
   (with $40.00 recordal fee)

☒ A Preliminary Amendment

☒ A copy of the International Search Report

☒ A copy of the Preliminary Examination Report

Fee Calculation:

☐ Applicant is entitled to Small Entity Status under 37 C.F.R. §1.27.

☒ BASIC FEE $860.00
(IPEA-U.S. $690/345; ISA-U.S. $710/355; PTO not ISA or IPEA $1000/500;
U.S. IPEA all claims meet 33(2)-(4) $100/50; File w/ EPO or JPO search report $860/430;)

☐ Surcharge for filing a late oath or declaration ($130/65) $

☐ Surcharge for filing a late translation ($130) $

☐ Assignment recordal fee ($40) $

☐ Multiple dependent claims ($270/135) $

☒ Excess claims - see calculation below $98.00

Total Claims:	21-	20 =	01	X $18/9 claim =	$18.00	
Independent Claims:	04-	3 =	01	X $80/40 ind. claim =	$80.00	
				Excess Claim Total	$98.00	

TOTAL FEES $958.00

☒ Check No.11805 in the amount of $958.00 is enclosed cover the filing fees (including the basic national fee under 37 CFR 1.492(a)) as calculated above.

☒ The Commissioner is authorized to charge any fees beyond the amount enclosed which may be required, or to credit any overpayment, to Deposit Account No. 500388 (Order No. UDL1P065).

General Authorization for Petition for Extension of Time (37 CFR §1.136)

☒ Applicants hereby make and generally authorize any Petitions for Extensions of Time as may be needed for any subsequent filings. The Commissioner is also authorized to charge any extension fees under 37 CFR §1.17 as may be needed to Deposit Account No. 500388 (Order No. UDL1P065).

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PATENT .TRADEMARK OFFICE

Date: _____ July 11, 2001 _____   C. _____

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of: PARVIN et al.	Attorney Docket No.: UDL1P065
Application No.: Not Yet Assigned	Examiner: Not Yet Assigned
Filed: Herewith	Group: Unknown

Title: APPARATUS AND METHODS FOR
INVESTIGATION OF RADIOACTIVE SOURCES
IN A SAMPLE

PRELIMINARY AMENDMENT

Commissioner for Patents
Washington, D.C. 20231

Dear Sir:

    Prior to examination on the merits, please enter the following preliminary amendment.

IN THE CLAIMS:

Please **AMEND** claims 3-5, 7-10, 15-17 and 19-20 as follows:

1.   A method of investigating radioactive sources in a sample, the method comprising detecting a portion of the emissions arising from the sample at an energy, the detected portion relating to a detected level, the detected level being corrected according to a correction method to give a corrected level, at that energy, the correction method including:-

the provision of an emission generator, passing at least a portion of the emissions of the generator into the sample, detecting at least a portion of the emissions from the generator leaving the sample, and determining a value for a first relationship between the two portions;

calculating a value for a relationship of equivalent type to the first, the calculation being based on functions of an element's absorption of emissions and the amount of that element potentially encountered by emissions, for one or more elements;

adjusting one or more variables / functions in the calculated relationship to reduce the difference between the value of the determined relationship and the value of the calculated relationship for the sample at a plurality of the energies of emissions from the generator; and

obtaining the values of the calculated relationship functions from the reduction and calculating the calculated relationship from those factors at the sample source emission energy requiring correction and correcting the detected level using those values.

2.   A method according to claim 1 in which the generator emissions are of at least two emission energies and at least two of those energies are detected.

3.   (Once Amended) A method according to claim 1 in which the calculated relationship is based on functions addressing one or more of the density, emission path length in the sample and sample absorption of emissions.

4. (Once Amended) A method according to claim 1 in which the calculated relationship is based on functions addressing one or both of the effect of the material forming the sample over the emission path length in the sample and the sample absorption of emissions.

5. (Once Amended) A method according to claim 1 in which the calculated relationship is based on the equation:-

$$T_i = \exp \quad -\sum q_j \cdot \mu_{i,j}$$

where T_i is the transmission coefficient at the energy i under consideration; q_j is the effective material thickness or the effect of the specified material forming the sample over the specified emission path length through the sample, for element j; $\mu_{i,j}$ is the mass absorption coefficient for elements j at energy i.

6.   A method according to claim 5 in which the calculated relationship includes contributions from three or more elements, j.

7. (Once Amended) A method according to claim 5 in which the elements include at least one low atomic mass element, preferably less than 10, at least one high atomic mass element, preferably greater than 40 and at least one intermediate atomic mass element, preferably between 10 and 50.

8. (Once Amended) A method according to claim 1 in which the adjusting of the variables / functions / factors varies one or two of the variables / functions / factors only.

9. (Once Amended) A method according to claim 1 in which the reduction in the differences between the first relationship value and calculated value is undertaken so as to reduce the overall difference between all of the first relationship and calculated relationship values involved.

10. (Once Amended) A method according to claim 1 in which the first relationship employs measured transmission coefficients.

11.   A method according to claim 10 in which the measured transmission coefficients, for one or more of the energies are provided according to the equation:-

$$\text{Trans. Coeff.} \quad = \quad \frac{R}{R_o}$$

where R is the rate of detected photons with the sample in place, R_o is the rate of photons which would be detected without the sample in place.

12.   Apparatus for investigating radioactive sources in a sample, the apparatus comprising:

one or more detectors for emissions from the sources, the detectors generating signals indicative of the emissions detected;

an investigating location into which the sample is introduced;

signal processing means for relating the detector signals to a detected level for the sources;

processing means for correcting the detected level for the sources, according to a correction method, to give a corrected level;

a radioactive emission generator separate from the sample;

one or more detectors for emissions from the generator
which leave the sample;

processing means for determining a first relationship,
based on the portion of generator emissions entering the sample
and the portion of generator emissions leaving the sample;

processing means for calculating a value for a relationship
of equivalent type to the first, the calculation being based on
functions of an element's absorption of emissions and the amount
of that element potentially encountered by emissions, for one or
more elements;

processing means for adjusting one or more variables in the
calculated relationship to reduce the difference between the
value of the determined relationship and the value of the
calculated relationship for the sample at a plurality of the
energies of emissions from the generator; and

calculating means for obtaining the values of the
calculated relationship functions from the reduction and
calculating the calculated relationship from those factors at
the sample source emission energy requiring correction and
correcting the detected level using that value.

13.   A method of investigating radioactive sources in a sample,
the method comprising detecting a portion of the emissions
arising from the sample, and further comprising the provision of
a radioactive generator, passing at least a portion of the
emissions of the generator into the sample, detecting at least a
portion of the emissions from the generator leaving the sample,
the radioactive generator emissions being of at least a
plurality of emission energies and at least two of those
energies being detected.

14.   A method according to claim 13 in which the emission
energies of the generator extend across a substantial portion of

the range of energies emitted from the sample, preferably a
substantial portion may be 50%.

15. (Once Amended) A method according to claim 13 in which the
generator emits energies encompassing the range of energies
emitted by the sample.

16. (Once Amended) A method according to claim 13 in which at
least 5 energies from the generator are detected and used.

17. (Once Amended) A method according to claim 13 in which the
method further provides that the detected portion of the source
emissions relate to a detected level for the sources in a
sample, the detected level being corrected according to a
correction method to give a corrected level for the sources in a
sample, the process being repeated for one or more other
samples.

18.   A method according to claim 17 in which the correction
method employs measured transmission coefficients in determining
the correction at the respective energies considered the
measured transmission coefficients, for one or more of the
energies being provided according to the equation:-

$$\text{Trans. Coeff.} \quad = \quad \frac{R}{R_0}$$

     where R is the rate of detected photons with the sample in
place, R_0 is the rate of photons which would be detected without
the sample in place.

19. (Once Amended) A method according to claim 17 in which for
correction of source emission energies corresponding to a

generator energy the measurement based correction factor for
that respective energy is used.

20. (Once Amended) A method according to claim 17 in which
correction of source emission energies not corresponding to a
generator energy is achieved using a correction factor based on
the extrapolation of the correction factors based on
measurements for two or more of the respective energies.

21. Apparatus for investigating radioactive sources in a
sample, the apparatus comprising:

     one or more detectors for emissions from the sources, the
detectors generating signals indicative of the emissions
detected;

     an investigating location into which the sample is
introduced;

     signal processing means for relating the detector signals
to one or more characteristics of the sources;

     a radioactive emission generator separate from the sample;
and

     one or more detectors for emissions from the radioactive
generator leaving the sample;

     wherein the radioactive generator emissions are of at least
a plurality of energies and a least two of the plurality of
energies are detected.

REMARKS

Claims 3-5, 7-10, 15-17 and 19-20 have been amended to remove multiple dependencies. Applicant believes that no additional fees are due upon filing this Amendment, however, should the Commissioner determine that additional fees are due, the Commissioner is hereby authorized to charge such fees to Deposit Account No. 50-0388.

Respectfully submitted,
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MARKED UP VERSION SHOWING CHANGES MADE

3. (Once Amended) A method according to claim 1 [or claim 2] in which the calculated relationship is based on functions addressing one or more of the density, emission path length in the sample and sample absorption of emissions.

4. (Once Amended) A method according to [any preceding] claim 1 in which the calculated relationship is based on functions addressing one or both of the effect of the material forming the sample over the emission path length in the sample and the sample absorption of emissions.

5. (Once Amended) A method according to [any preceding] claim 1 in which the calculated relationship is based on the equation:-

$$\mathbf{T_i = exp} \quad -\sum \mathbf{q_j} \cdot \mu_{i,j}$$

where T_i is the transmission coefficient at the energy i under consideration; q_j is the effective material thickness or the effect of the specified material forming the sample over the specified emission path length through the sample, for element j; $\mu_{i,j}$ is the mass absorption coefficient for elements j at energy i.

7. (Once Amended) A method according to claim 5 [or claim 6] in which the elements include at least one low atomic mass element, preferably less than 10, at least one high atomic mass element, preferably greater than 40 and at least one intermediate atomic mass element, preferably between 10 and 50.

8. (Once Amended) A method according to [any preceding] claim 1
in which the adjusting of the variables / functions / factors
varies one or two of the variables / functions / factors only.

9. (Once Amended) A method according to [any preceding] claim 1
in which the reduction in the differences between the first
relationship value and calculated value is undertaken so as to
reduce the overall difference between all of the first
relationship and calculated relationship values involved.

10. (Once Amended) A method according to [any preceding] claim
1 in which the first relationship employs measured transmission
coefficients.

15. (Once Amended) A method according to claim 13 [or claim 14]
in which the generator emits energies encompassing the range of
energies emitted by the sample.

16. (Once Amended) A method according to [any of claims] **claim**
13 [to 15] in which at least 5 energies from the generator are
detected and used.

17. (Once Amended) A method according to [any of claims] **claim**
13 [to 16] in which the method further provides that the
detected portion of the source emissions relate to a detected
level for the sources in a sample, the detected level being
corrected according to a correction method to give a corrected
level for the sources in a sample, the process being repeated
for one or more other samples.

19. (Once Amended) A method according to claim 17 [or claim 18]
in which for correction of source emission energies
corresponding to a generator energy the measurement based
correction factor for that respective energy is used.

20. (Once Amended) A method according to [any of claims] **claim** 17 [to 19] in which correction of source emission energies not corresponding to a generator energy is achieved using a correction factor based on the extrapolation of the correction factors based on measurements for two or more of the respective energies.

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APPARATUS AND METHODS FOR INVESTIGATION OF RADIOACTIVE SOURCES IN A SAMPLE

This invention is concerned with improvements in and relating to apparatus and methods for materials investigations. The invention is particularly, but not exclusively, concerned with investigating gamma ray emissions from materials. The invention is still more particularly, but not exclusively, concerned with correction techniques including the provision of more suitable transmission sources and an improved correction technique.

In a variety of situations it is necessary to investigate emissions from radioactive sources in or on materials to form a basis for a variety of subsequent decisions, actions or further considerations. The investigations of the samples may relate directly to the emission, for instance the emission source, or indirectly, for instance the consideration of associated non-emitters or emitters which are not directly measurable. The emissions of interest are in particular gamma ray emissions, but other emission forms may be considered additionally or alternatively.

Emission investigation is particularly important in waste evaluation cases. For a given waste sample it is desirable to be able to determine a variety of unknowns. The unknowns may include, but are not limited to, one or more of the level, type, constituents, nature and distribution of the emissions, emission sources, associated materials or associated factors.

As well as simply using the results obtained from the detectors, emission count rates for the radioactive materials in the sample under consideration, it is known to investigate to an extent the effects of the all the material in a sample, through their effect on the transmission of external

emissions from an applied source through the sample to the
detectors.

The applicant has determined that such simple sources
are not suitable for transmission based investigation and
correction at the full variety of energies and with the full
variety of materials. As a consequence the effectiveness of
the correction technique based on these investigations is
impaired.

The present invention seeks to provide a more widely
accurate transmission based correction, which is also simple
and easy to use across a full spread of situations which are
encountered in practice.

In addition the existing transmission based correction
techniques only go part way to account for the variables
which affect the detected counts for the sources of the
sample itself and as a result the correction technique is not
fully effective.

The present invention has amongst its aims the
provision of a more accurate and thorough correction
technique based on transmission correction and the manner in
which it is employed.

According to a first aspect of the invention we provide
a method of investigating radioactive sources in a sample,
the method comprising detecting a portion of the emissions
arising from the sample, and further comprising the provision
of a radioactive generator, passing at least a portion of the
emissions of the generator into the sample, detecting at
least a portion of the emissions from the generator leaving
the sample, the radioactive generator emissions being of at
least a plurality of emission energies and at least two of
those energies being detected.

Preferably the method further provides that the
detected portion of the source emissions relate to a detected

-3-

level for the sources in a sample, the detected level being
corrected according to a correction method to give a
corrected level for the sources in a sample, the process
being repeated for one or more other samples.

Preferably the correction method employs measured
transmission coefficients in determining the correction. The
measured transmission coefficients, for one or more of the
energies, most preferably all, may be provided according to
the equation :-

$$\text{Trans. Coeff.} = \frac{R}{R_o}$$

where R is the rate of detected photons with the sample
in place, R_o is the rate of photons which would be detected
without the sample in place.

Preferably the density determined is used as a factor
in the correction method. The density used in the correction
method may be an averaged density from the determinations or
a weighted average density from the determinations.

For correction of source emission energies
corresponding to a generator energy preferably the
measurement based correction factor is used. For correction
of source emission energies not corresponding to a generator
energy preferably the correction factor is based on the
extrapolation of the correction factors based on the
measurements.

The generator is preferably a single isotope.
Preferably the emission energies extend across a substantial
portion of the range of energies emitted from the sample. A
substantial portion may be 50%, preferably 75%, more
preferably 90% and ideally 100% of the sample energies range.
The generator most preferably of all emits energies
encompassing the range of energies emitted by the sample.

−4−

^{152}Eu is a particularly preferred generator. Preferably at least 5 energies from the source are detected and used, more preferably at least 8 energies are detected and used.

Preferably the portion of generator emissions detected have passed through the sample. Preferably the generator is provided on the opposing side of the sample to the detectors, most preferably in direct opposition.

One or more of the detectors for the sources may be used for detecting the generator emissions and/or vice-versa.

According to a second aspect of the invention we provide apparatus for investigating radioactive sources in a sample, the apparatus comprising :

one or more detectors for emissions from the sources, the detectors generating signals indicative of the emissions detected;

an investigating location into which the sample is introduced;

signal processing means for relating the detector signals to one or more characteristics of the sources;

a radioactive emission generator separate from the sample; and

one or more detectors for emissions from the radioactive generator leaving the sample;

wherein the radioactive generator emissions are of at least a plurality of energies and a least two of the plurality of energies are detected.

The source detectors and the generator detectors may be one and the same in the case of one or more or all of the detectors.

The second aspect of the invention may include any of the features, options and possibilities set out on the first aspect of the invention, including apparatus suitable for the implementation of the method steps detailed therein.

-5-

According to a third aspect of the invention we provide a method of investigating radioactive sources in a sample, the method comprising detecting a portion of the emissions arising from the sample at an energy, the detected portion relating to a detected level, the detected level being corrected according to a correction method to give a corrected level, at that energy, the correction method including :-

the provision of an emission generator, passing at least a portion of the emissions of the generator into the sample, detecting at least a portion of the emissions from the generator leaving the sample, and determining a value for a first relationship between the two portions;

calculating a value for a relationship of equivalent type to the first, the calculation being based on functions of an element's absorption of emissions and the amount of that element potentially encountered by emissions, for one or more elements;

adjusting one or more variables / functions in the calculated relationship to reduce the difference between the value of the determined relationship and the value of the calculated relationship for the sample at a plurality of the energies of emissions from the generator; and

obtaining the values of the calculated relationship functions from the reduction and calculating the calculated relationship from those factors at the sample source emission energy requiring correction and correcting the detected level using those values.

Preferably the generator emissions are of at least two emission energies, ideally multi-energy emissions are provided, and / or with at least two of those energies being detected.

The generator is preferably a single isotope. Preferably the emission energies extend across a substantial

-6-

portion of the range of energies emitted from the sample. A
substantial portion may be 50%, preferably 75%, more
preferably 90% and ideally 100% of the sample energies range.
The generator most preferably of all, emits energies
encompassing the range of energies emitted by the sample.
Eu^{152} is a particularly preferred generator.

Preferably the portion of generator emissions detected
have passed through the sample. Preferably the generator is
provided on the opposing side of the sample to the detectors,
most preferably in direct opposition.

One or more of the detectors for the sources may be
used for detecting the generator emissions and/or vice-versa.

Preferably the first relationship employs measured
transmission coefficients, and ideally comprises measured
transmission coefficients. The measured transmission
coefficients, for one or more of the energies, most
preferably all, may be provided according to the equation :-

$$\text{Trans. Coeff.} \quad = \quad \frac{R}{R_o}$$

where R is the rate of detected photons with the sample
in place, R_o is the rate of photons which would be detected
without the sample in place.

Preferably the calculated relationship is based on
functions addressing one or more of the density, emission
path length in the sample and sample absorption of emissions.
Preferably the calculated relationship is based on functions
addressing one or both of the effect of the material forming
the sample over the emission path length in the sample and
the sample absorption of emissions.

The calculated relationship is preferably based on the
equation :-

–7–

$$T_i = \exp\left(-\sum q_j \cdot \mu_{i,j}\right)$$

where T_i is the transmission coefficient at the energy i
under consideration; q_j is the effective material thickness
or the effect of the specified material forming the sample
over the specified emission path length through the sample,
for element j; $\mu_{i,j}$ is the mass absorption coefficient for
elements j at energy i. The sum including all of the
specified elements, j, included in the method.

Preferably the calculated relationship includes
contributions from two or more elements, and most preferably
three or more. The elements may be elements in the sample,
likely to be in the sample, unknown or not in the sample.

Most preferably the elements include at least one low
atomic mass element, preferably less than 10 and ideally less
than 5. Preferably the elements include at least one high
atomic mass element, preferably greater than 40 and ideally
greater than 50. Preferably the elements include at least
one intermediate atomic mass element, preferably between 10
and 50 and ideally between 10 and 40. Ideally at least one
element from each category is provided.

Preferably the adjusting of the variables / functions /
factors varies one or two of the factors only. Preferably
only the effective material thickness or the effect of the
specified material forming the sample over the specified
emission path length through the sample are varied,
particularly where one factor only is varied.

The reduction in the differences between the first
relationship value and calculated value may be undertaken so
as to reduce the overall difference between all of the first
relationship and calculated relationship values involved.
The reduction may be intended to minimise the difference.
The reduction process may be a minimisation of the sum of the
residuals.

-8-

Preferably the method is repeated for one or more other samples, the one or more other samples may, with the sample, form a body of material under investigation.

For the correction of source emission energies corresponding to a generator emission energy, preferably the measurement based correction factor alone, i.e. the first relationship, is used.

According to a fourth aspect of the invention we provide apparatus for investigating radioactive sources in a sample, the apparatus comprising :

one or more detectors for emissions from the sources, the detectors generating signals indicative of the emissions detected;

an investigating location into which the sample is introduced;

signal processing means for relating the detector signals to a detected level for the sources;

processing means for correcting the detected level for the sources, according to a correction method, to give a corrected level;

a radioactive emission generator separate from the sample;

one or more detectors for emissions from the generator which leave the sample;

processing means for determining a first relationship, based on the portion of generator emissions entering the sample and the portion of generator emissions leaving the sample;

processing means for calculating a value for a relationship of equivalent type to the first, the calculation being based on functions of an element's absorption of emissions and the amount of that element potentially encountered by emissions, for one or more elements;

processing means for adjusting one or more variables in
the calculated relationship to reduce the difference between
the value of the determined relationship and the value of the
calculated relationship for the sample at a plurality of the
energies of emissions from the generator; and

calculating means for obtaining the values of the
calculated relationship functions from the reduction and
calculating the calculated relationship from those factors at
the sample source emission energy requiring correction and
correcting the detected level using that value.

The processing means may be separate from one another
or a common unit in one or more cases, including the
calculating means.

The fourth aspect of the invention may include any of
the features, options and possibilities set out on the third
aspect of the invention, including apparatus suitable for the
implementation of the method steps detailed therein.

The first and/or second and/or third and/or fourth
aspects of the invention may further include any of the
features, options, possibilities and steps set out below.

The sources may be singular or plural in disposition
and/or type. The sources may be one or more isotopes of one
or more elements. The sources may be alpha and/or beta
and/or gamma emitters, but are preferably gamma emitters at
least.

One or more sources of the same type and/or of
different types may be present in the sample. The sources
may be homogeneously distributed, or more usually, unevenly
distributed. The size and/or shape and/or mass of a source
may be different from the size and/or shape and/or mass of
another source in the sample, be they of the same or
different types.

The sources may be investigated by detecting one or more of their emitting energies. Thus a characteristic energy of an isotope may be detected.

The sources may be investigated directly, for instance they contribute directly to the detected level, and/or the sources may be investigated indirectly, for instance they do not contribute directly to the detected level but are associated with sources which do.

The samples may be gaseous and/or liquid and/or solid. The samples may contain one or more non-emitting or non-source materials. The materials may include one or more of metals, such as iron, steel, aluminum; wood; glass; plastics, such as polythene, PVC; liquids, such as water.

The sample may be the whole or part of a body of material. The body of material may be free standing, but is preferably contained in a container. The sample may be a part of a body of material, including the part of the container associated with that part of the body of material.

The sample may be a segment or slice through a body of material. Preferably the segment is taken horizontally through the body of material. Preferably the segment has the same thickness throughout the body of material.

Other segments of the body of material may be investigated in subsequent repeats of the method.

The container preferably entirely encloses the body of material. The container may be of metal or of concrete or a combination of such materials. Drums are a particularly preferred container, such as right cylindrical drums.

The containers may be of one or more standard sizes. The height and/or diameter of the containers may be standard.

Preferably the containers introduced to the investigating location one at a time. The containers may be introduced by conveying along a surface, preferably a horizontal surface. The surface may include or be formed of a plurality of rollers. Preferably the container is removed

from the investigating location in a manner equivalent to its
introduction.

The investigating location is preferably provided in
proximity to the emission detector or detectors. The
investigating location may be provided in proximity to one or
more radioactive sources. The sources are preferably
intended to transmit radiation through the sample. Ideally
the investigating location is provided between the
detector(s) and the transmission source(s).

The sample, preferably the container for it, may be
rotated at the investigating location. Preferably the
rotation presents different portions of the sample in
proximity to the detector(s) and/or transmission source(s).
the rotation may be continuous or stepped. The rotation may
be provided at between 5 and 25 rpm.

Preferably the sample and/or body of material and/or
container are weighed, most preferably at the investigating
location, for instance by the turntable used to rotate it.

The sample, preferably the container for it, may be
raised and/or lowered at the investigating location.
Preferably the rasing and/or lowering presents different
portions of the sample to the detector(s) and/or transmission
source(s). the raising and/or lower may be continuous or
stepped. Preferably investigations are performed as the
sample is lower and raised.

The sample may be rotated and/or lower and/or raised.

A single detector may be used. Preferably a plurality
of detectors, for instance three, may be used.

The detectors may be of the high purity germanium type.

Preferably the detectors are collimated to restrict
their field of view to the body of material of which the
sample is the whole or a portion thereof. Where the sample
is less than the whole of the body of material, preferably
the detectors are collimated to restrict their field of view
to the sample only. The sample is preferably a slice or

-12-

segment of the whole. The segments may be of the same
thickness.

     Preferably the detected level is obtained from a
passive counting stage. Preferably the transmission based
investigations are performed before and/or after the passive
count stage.

     Preferably the transmission source(s) is provided in
opposition to the detector(s). Preferably the same number of
transmission sources are provided as there are detectors. It
is particularly preferred that the transmission source be
provided according to nature of the transmission source
detailed in the first and/or second aspect of this invention.

     One or more surface dosimeters may be provided.
Preferably the surface dosimeters are configured.to
investigate gamma emitting sources. Alpha and/or beta
emissions may alternatively or additionally considered.

     Preferably the correction method is used in correcting
the detected level to the corrected level, for instance by
established subsequent techniques, such as those set out in
the Los Alamos primer, 2nd Edition, March 1991, ISBN0-16-
032724-5.

     Various embodiments of the invention will now be
described, by way of example only, and with reference to the
accompanying drawings in which :-

           _   Figure 1 illustrates schematically an instrument
    suitable for implementing the present invention;

               Figure 2 illustrates measured and fitted
    transmission coefficients calculated according to one
    embodiment of the invention for a drum containing a paper
    matrix (density <0.1g.cm^{-3});

               Figure 3 illustrates measured and fitted
    transmission coefficients for a PVC matrix (density =
    0.4g.cm^{-3}); and

–13–

Figure 4 illustrates measured and fitted
transmission coefficients for an iron matrix (density
$1.5g.cm^{-3}$).

The instrument illustrated in Figure 1 is suitable for
investigating gamma emission sources in a variety of
situations and materials. The system 1 is particularly
designed to investigate waste samples presented in drums 3 to
investigating location 5.

The drums considered may be in a variety of sizes, but
the instrument is readily adapted to consider 100, 200 and
500 litre drums. Masses of waste, such as 40kg to 550kg, can
readily be accommodated.

The drums 3 may be provided with a barcode which can be
examined by a barcode reader on the system 1 so as to
permanently assign results obtained to that drum 3.

The system 1 features a conveying table 7 formed of a
large number of parallel rollers 9, onto which the drums are
lowered. Cranes, forklift trucks and other lifting and
manoeuvring means can be used to this end.

The conveying table 7 leads to the investigating
location 5 and then onward out the other side to a
dismounting location 11 from which the drums 3 are lifted.
This set up allows the flow of samples for investigation
through the system.

The drum 3 is supported at the investigating location 5
by a turntable 13 which again is formed by a number of
parallel rollers 15 mounted on a moveable frame 17. The
frame 17 can be rotated about a vertical axis, using a motor
(not shown), so as to rotate the drum 3 about its
longitudinal axis at the investigating location 5. A
rotational rate of 12 rpm is preferred, but stepped rotation
can be used. The frame 17 can also be provided with the
option of being raised and lowered relative to the level of
the surrounding conveying table 7, using electrical drives,

-14-

hydraulics or other systems (not shown), so as to adjust the
vertical positioning of the drum 3 within the investigating
location 5. The lowering and rotating motions may be applied
simultaneously.

The turntable 13 and frame 17 are configured to weigh
the drum 3 whilst it is on the turntable 13.

On either side of the investigating location 5 are
investigating assemblies 19, 21. In general one of these
assemblies 19 acts as the, optional, transmission side and
the other 21 as the detecting/receiving side for the
investigations.

The detecting/receiving side 21 provides one or more
detectors 23 for the emission type under consideration. The
detectors 23 are collimated using shields 25 to give a
restricted field of view into the investigating location 5
for each detector 23. Through the use of variable aperture
collimators (not shown) the range of radioactivity level for
the waste which can successfully be handled is increased.
The field of view is generally configured to be a slice
through that investigating location 5, the slice being
substantially parallel to the turntable 13 and/or
perpendicular to the axis of the drum 3 under consideration.

For gamma rays the detectors may be of the germanium
type, for instance high purity germanium type. LRGS or HRGS
detectors may be used, with electrical or LN_2 cooling for
HRGS detectors.

The provision of more than one detector, collimated to
different fields of view, allows a greater number of
measurements to be taken simultaneously, hence increasing the
throughput for the system.

The detectors 23 monitor gamma rays originating in
their field of view in the drum 3 and in effect generate
count rates. A count time of less than 30 minutes is
generally employed.

-15-

The signals obtained from the detectors are fed to processing electronics 27 and hence to a CPU 29 and operator display 31. Operator control and inputs are facilitated through keyboard 33.

The processing electronics 27 are provided with error handling functions and diagnostic facilities, as well as providing the appropriate calibration functions.

Processing of the signals gives detailed information on the assay of waste material in the drum 3, the isotopic make-up of the waste. More details of these analyses are discussed below. The results can be used to classify waste according to the relevant disposal categories, including those below a deminimus level which can be characterized as non-radioactive. The results can be expressed as the identification of fission products, activation products or MGA code. The results can also be combined with the "fingerprinting" technique to give non-measurable isotope determinations.

The results obtained can be improved using a variety of potential correction techniques. Correction based on weight and/or differential peak adsorption and/or use of a transmission source may be used. In this invention the new correction techniques detailed in the same applicant's UK Patent Application no. 9900448.3 designated P17453 filed on 11 January 1999 may also be used and details of that technique are fully incorporated herein by reference. This new technique is based on considering the contents of the drum as a number of distinct parts, for instance horizontal segments through the drum, each of the segments being treated separately from the others in terms of its density value. By treating the drum as a number of potentially different density parts the errors referred to above are substantially removed. This division of the drum into segments and allocation of a density value to each segment cannot be made based on the weight of the drum alone.

-16-

To achieve the aim the technique evaluates the amount
of material in a given segment by means of transmission
source based investigations.

If R_o is the rate at which photons from an external
source are detected in the absence of the drum and R is the
rate of detection of those photons with the drum present
then :-

$$R = R_o \exp(-\mu\rho x)$$

where μ is the mass attenuation coefficient, ρ is the
density of the sample and x is the thickness of the matrix.

For any given drum x will be constant as this is the
path length through the drum (56cm for a 200 litre drum, for
instance). At high gamma energies, say greater than 400keV,
μ can be treated as effectively constant too. Experimental
demonstration of this is provided below. In effect these
valid assumptions make ρ proportional to $-\ln(R/R_o)$.

For a given segment or slice, s, through the drum, the
amount of material in that slice, V_s, is assigned the value
$-\ln(R/R_o)$. The total amount of the material present in the
drum is thus defined as proportional to $\sum V_s$; the sum being
for all the slices of the drum.

Following this definition, the fraction of the whole
body of material in a given slice, s_1, is given by $V_{s1}/\sum V_s$.
As the total mass of the drum is known (from weighing), as
the total volume of the drum is known (from measurement or as
a standard size) and as the number of segments into which the
drum has been divided is known (from the analysis control),
then the density is defined as :-

$$\rho = (N.M/V) . (V_s/\sum V_s)$$

−17−

The provision of more accurate density information specific to each slice allows more accurate attenuation correction for that slice and hence greater overall accuracy.

Through the use of a transmission source or sources 35 on transmission side 19 in combination with stepped rotation of the drum 3 tomographic style investigations of the drum can be made to give plots of density distribution and radioactive distribution for the drum 3.

Surface originating alpha and/or beta emissions, most preferably gamma emissions, for the drum 3 can also be measured using optional dosimeters 37. The dosimeters are normally provided on the transmission side 19 in proximity with the surface of the drum 3.

As discussed above three different known correction techniques may be deployed. The principles and operation behind each of these is now discussed.

Weight based correction seeks to account for the attenuating effects of the body of material the sources are in by a factor based on the body of materials density. The total mass of the entire body of material is divided by the total volume of the body of material, more commonly the total volume of the container for the body, to give an overall density value for the entire body/container. This single value is then used in the correction of the detected count to account for the reduction arising due to attenuation.

Differential peak adsorption based correction again seeks to account for the attenuation effects of the material, but through a more direct investigation of attenuation. The gamma emissions from a source anticipated to be in the body of material at two characteristic energies are considered. The ratio of the emissions at one energy to the emissions at the other energy is known (1:1, for instance, for Co-60) without attenuating materials, and this base ratio is compared with the actual ratio measured with the attenuating effects of the body of material present to give a factor

relating to the attenuating effect and hence allowing
correction.

Transmission source based correction comes in a variety
of forms, but each is generally based on determining
attenuation effects on emissions from within the body of
material by measuring the attenuation on externally sourced
gamma emissions. Emissions at a known energy from the
transmission source are detected after their passage through
the body. The ratio of the detected emissions with the body
of material present is compared with the detected emissions
which would occur without the material. The attenuation is
corrected for based on this difference. The energy of the
transmission source is selected to be close to the energy of
the emissions from the sample which need correcting.

The prior art transmission correction based techniques
present a number of problems or inaccuracies in practice.

The use of a mono-energetic transmission source, as has
been deployed to date, renders the correction technique
effective to varying degrees for varying waste or sample
materials.

The samples under consideration may contain isotopes
which emit at a variety of different energies within the
spectrum. The energies can be quite low in some instances
and high for other instances or be spread across the
substantial part of the spectrum.

The mono-energetic sources used to date have been used
to determine a correction factor according to the measured
effect of the sample on the detected signal for that source
following transmission through the sample. The correction
factor has been used in relation to all the detected
emissions from the sample itself, irrespective of their
energy.

In reality the applicant has established that the
information gained at that particular energy (and hence the
correction factor calculated) are not appropriate across the

−19−

full width of the spectrum or even a reasonable part thereof.
Once away from energies the same as or close to the energy of
the mono-energetic source the correction factor becomes
inaccurate. Furthermore, the extent and manner of variation
of the effects of the sample vary across the spectrum
according to the sample characteristics too and as a
consequence are not predictable in advance.

        To address the problem the present invention employs a
multi-energetic source as the transmission source for sample
investigation. The sources used are carefully selected to
provide energies spanning the important part of the spectrum
for a number of commonly encountered waste types. The source
material is exemplified by ^{152}Eu. The intention is to provide
a series of transmission based investigations which bracket
the emissions from within the sample itself. Thus a more
appropriate correction factor can be calculated for the
sample in question and its actual emission energies as the
deviation of those energies from the energies at which the
transmission effects are actually measured and known are
significantly reduced.

        The overall effect of the multi-energy source is that
the correction is more accurate and the likely error is
reduced.

        The actual correction is obtained from the measured
transmission coefficients, which at the respective energies
are :-

$$\text{Trans. Coeff.} \quad = \quad \frac{R}{R_o} \quad = \exp(-\mu\rho x)$$

        where R is the rate of detected photons, R_o is the rate
of emitted photons from the source, μ is the mass absorption
coefficient, ρ is the matrix density, x is the matrix
thickness.

-20-

Measured transmission coefficients for two different
samples with significantly different make-ups, at the various
energies given by ^{152}Eu, would give rise to significant
variation with energy between the two samples, a variation
which would not be apparent from a single investigation at
using transmission correction based on a single energy.

Between the greater number of actually measured values
provided by the new technique, an extrapolated value can be
used. The greater number and range of the measured values
make this extrapolation more accurate too.

Over and above this correction technique, which is
fully accurate at more points and more accurate at the
intervening locations, the present invention offers an
optimized technique which makes the correction fully
effective at all energies.

As well as addressing the range of energies for which
the correction is accurate, the enhanced technique also
addresses addition problems with the correction techniques
applied in the past, as they only went part way to addressing
the variations in sample material. The wastes and other
samples which need to be investigated vary significantly
between batches of waste and even between individual drums.
The waste may be soft waste (approx. 0.1g.cm^{-3}) or metallic
and/or ashes and/or cement containing and/or compacted waste
(approx. 2.0g.cm^{-3} or even greater). These variations have
significant effects, but the present technique accounts for
the unknown material composition to a very large part too.

In addition to the measured transmission coefficients
obtained as described above, the technique also deploys
fitted transmission coefficients.

In this technique the material forming the sample is
assumed to be made up of three or more elements in unknown
ratios. The transmission coefficient at energy i is the sum
of the exponential terms for each of the constituents, i.e.
for j terms. The definition is :-

$$T_{nj} = \exp\left(-\sum_{j} q_j u_{ij} \right)$$

where q_j is the effective matrix thickness for material j and μ_{ji} is the mass absorption coefficient for material j at energy i.

Three or more materials can be used in the determination, but it is preferred that one low atomic number, one mid atomic number and one high atomic number constituent at least be used (i.e. low Z, mid Z and high Z elements). The elements may be hydrogen (low), aluminum (mid) and iron (high), for instance. It should be noted that the material selected need not be a constituent of the sample for the technique to work.

Using this formula transmission coefficients can be calculated by varying the q_j values. The variations are aimed at minimizing the sum of the residuals from a comparison of the measured and calculated/fitted coefficients. The fitting may be a linear least squares approach (matrix solution) or cycling through the possible q values. Once minimized a set of q_j values are reached which can be used to calculate accurately the transmission coefficient at any desired energy i.

In effect the technique assigns a fixed proportion of each composite material to best describe the unknown elemental composition of the sample. The result is an accurate transmission coefficient at any energy and hence full correction at any energy.

Figure 2 illustrates measured and fitted transmission coefficients calculated in the above mentioned manner for a 200 litre drum containing a paper matrix (density <0.1g.cm⁻³), whilst Figure 3 illustrates measured and fitted transmission coefficients for a PVC matrix (density = 0.4g.cm⁻³) and Figure 4 illustrates measured and fitted

-22-

transmission coefficients for an iron matrix (density
1.5g.cm^{-3}).

     This additional correction technique offers accurate
correction over a wide energy range and also avoids the need
for the transmission source to have emission energies which
bracket the energy of the isotope of interest (without loss
of accuracy) as correction can accurately be extrapolated and
compositional assumptions on the sample material are not
needed.

     The techniques described above can be used for a
container or drum taken as a whole. However, the technique
is adapted to a more accurate and thorough investigation
which takes into account likely variations between different
parts of the sample.

     The technique, improved measured transmission based
correction and/or the still more accurate measured and fitted
transmission based correction can both be applied to
individual slices or other parts of the sample/container by
using suitable collimated detector fields of view. In this
way density variations in the sample with, for instance
height, can be accounted for. Similarly variations in
material composition with, for instance height, can be
accounted for and partially filled drums can be handled
accurately. No assumptions on the compositional make up of
the material or its distribution need be made.

     The results of the individual parts can be considered
separately or in combination to give an overall view point.

CLAIMS:

1.   A method of investigating radioactive sources in a
sample, the method comprising detecting a portion of the
emissions arising from the sample at an energy, the detected
portion relating to a detected level, the detected level
being corrected according to a correction method to give a
corrected level, at that energy, the correction method
including:-

      the provision of an emission generator, passing at
least a portion of the emissions of the generator into the
sample, detecting at least a portion of the emissions from
the generator leaving the sample, and determining a value
for a first relationship between the two portions;

      calculating a value for a relationship of equivalent
type to the first, the calculation being based on functions
of an element's absorption of emissions and the amount of
that element potentially encountered by emissions, for one
or more elements;

      adjusting one or more variables / functions in the
calculated relationship to reduce the difference between the
value of the determined relationship and the value of the
calculated relationship for the sample at a plurality of the
energies of emissions from the generator;   and

      obtaining the values of the calculated relationship
functions at a reduced difference, and calculating from
those function valves, the calculated relationship at the
sample source emission energy requiring correction, the
detected level being corrected to the corrected level using
those function values.

2.   A method according to claim 1 in which the generator
emissions are of at least two emission energies and at least
two of those energies are detected.

3.   A method according to claim 1 or claim 2 in which the calculated relationship is based on functions addressing one or more of the density, emission path length in the sample and sample absorption of emissions.

4.   A method according to any preceding claim in which the calculated relationship is based on functions addressing one or both of the effect of the material forming the sample over the emission path length in the sample and the sample absorption of emissions.

5.   A method according to any preceding claim in which the calculated relationship is based on the equation:-

$$T_i = \exp\left(-\sum q_j . \mu_{i,j}\right)$$

where T_i is the transmission coefficient at the energy i under consideration; q_j is the effective material thickness or the effect of the specified material forming the sample over the specified emission path length through the sample, for element j; $\mu_{i,j}$ is the mass absorption coefficient for elements j at energy i.

6.   A method according to claim 5 in which the calculated relationship includes contributions from three or more elements, j.

7.   A method according to claim 5 or claim 6 in which the elements include at least one low atomic mass element, preferably less than 10, at least one high atomic mass element, preferably greater than 40 and at least one

intermediate atomic mass element, preferably between 10 and 50.

8.   A method according to any preceding claim in which the adjusting of the variables / functions / factors varies one or two of the variables / functions / factors only.

9.   A method according to any preceding claim in which the reduction in the differences between the first relationship value and calculated value is undertaken so as to reduce the overall difference between all of the first relationship and calculated relationship values involved.

10.   A method according to any preceding claim in which the first relationship employs measured transmission coefficients.

11.   A method according to claim 10 in which the measured transmission coefficients, for one or more of the energies are provided according to the equation:-

$$Trans.\ Coeff. = \frac{R}{R_o}$$

where R is the rate of detected photons with the sample in place, R_o is the rate of photons which would be detected without the sample in place.

12.   A method according to any of claims 1 to 11 in which the generator emits energies encompassing the energy or range of energies emitted by the sample.

-26-

13. A method according to any of claims 1 to 11 in which the emission energies of the generator extend across at least 50% of the range of energies emitted from the sample.

14. A method according to any of claims 1 to 13 in which at least 5 energies from the generator are detected and used.

15. Apparatus for investigating radioactive sources in a sample, the apparatus comprising:

one or more detectors for emissions from the sources, the detectors generating signals indicative of the emissions detected;

an investigating location into which the sample is introduced;

signal processing means for relating the detector signals to a detected level for the sources;

processing means for correcting the detected level for the sources, according to a correction method, to give a corrected level;

a radioactive emission generator separate from the sample;

one or more detectors for emissions from the generator which leave the sample;

processing means for determining a first relationship, based on the portion of generator emissions entering the sample and the portion of generator emissions leaving the sample;

processing means for calculating a value for a relationship of equivalent type to the first relationship, the calculation being based on functions of an element's absorption of emissions and the amount of that element potentially encountered by emissions, for one or more elements;

processing means for adjusting one or more values of the functions in the calculated relationship to reduce the
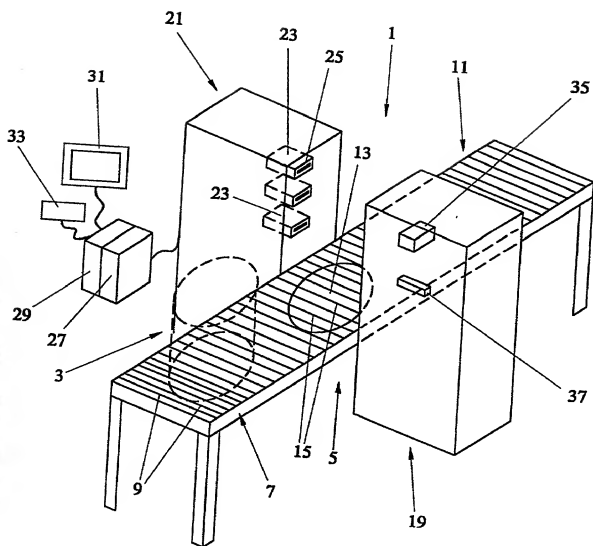
difference between the value of the determined relationship
and the value of the calculated relationship for the sample
at a plurality of the energies of emissions from the
generator; and

calculating means for obtaining the values of the
calculated relationship functions at the reduced difference
and calculating from those function values the calculated
relationship at the sample source emission energy requiring
correction, the detected level being corrected to the
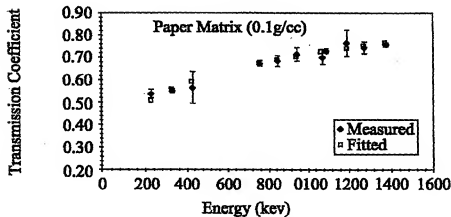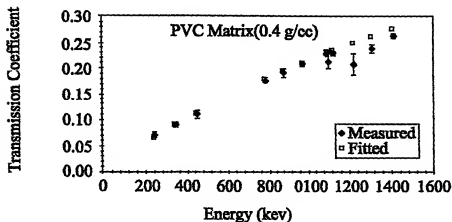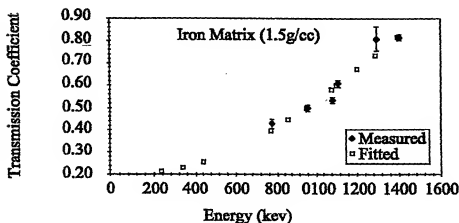corrected level using those values.

FIG. 1

-2/2-

FIG.   2

FIG.   3

FIG.   4

DECLARATION AND POWER OF ATTORNEY
FOR ORIGINAL U.S. PATENT APPLICATION

Attorney's Docket No. UD&LP065

As a below-named inventor, 1 hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe that I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled: APPARATUS AND METHODS FOR INVESTIGATION OF RADIOACTIVE SOURCES IN A SAMPLE the specification of which,

(check one)   1. ☐   is attached hereto.

                     2. ☐   was filed on _____ as
                             U.S. Application No. _____
                             and was amended on _____ .

                     3. ☒   was filed on January 11, 2000 _____ as
                             International PCT Application No. _PCT/GB00/00009_____
                             and was amended on _____ .

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the patentability of this application in accordance with Title 37, CFR § 1.56.

Prior Foreign Application(s)

I hereby claim foreign priority benefits under Title 35, United States code, § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed:

			Priority Benefits Claimed?
9900449.1	Great Britain	January 11, 1999	Yes _X_ No __
(Application No.)	(Country)	(Filing Date)	
9900448.3	Great Britain	January 11, 1999	Yes _X_ No __
(Application No.)	(Country)	(Filing Date)	

Provisional Application(s)

I hereby claim the benefit under 35 U.S.C. §119(e) of any United States provisional application(s) listed below:

_____	_____
(Application No.)	(Filing Date)

_____	_____
(Application No.)	(Filing Date)

Prior U.S. Application(s)

I hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s), or § 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of Title 35, United States Code, § 112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application:

(Application No.)	(Filing Date)	(Status - patented, pending, abandoned)

(Application No.)	(Filing Date)	(Status - patented, pending, abandoned)

Power of Attorney

And I hereby appoint the law firm of Beyer Weaver & Thomas, LLP and all practitioners who are associated with the Customer Number 022434 as my principal attorneys to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith.

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

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